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The effect of X-ray irradiation on ultrasound attenuation and velocity in LiF single crystals

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Using the pulsed method at a frequency of 7,5 MHz, the dependences of dislocation absorption and ultrasound velocity in LiF single crystals with residual deformation ε = 0,65 % at T = 300 K in the dose range of 0–1057 R were investigated. It has been established that, under conditions of crystals' irradiation with X-rays, the concentration of radiation centers fixing easily mobile dislocations significantly increases. As a result of limiting these dislocations' number, the attenuation of ultrasound decreases sharply, and its propagation velocity in the sample increases.
Keywords: irradiation, ultrasound attenuation and velocity, density of highly mobile dislocations, average effective length of a dislocation segment, deformation, radiation pinning centers.

Импульсным методом на частоте 7,5 МГц исследованы зависимости дислокационного поглощения и скорости ультразвука в монокристаллах LiF с остаточной деформацией ε = 0,65 % при T = 300 К в интервале доз облучения 0–1057 рентген. Установлено, что в условиях облучения кристаллов рентгеновскими лучами существенно увеличивается концентрация радиационных центров, закрепляющих легкоподвижные дислокации. В результате ограничения числа указанных дислокаций затухание ультразвука резко снижается, а скорость его распространения в образце увеличивается.

Вплив рентгенівського опромінення на згасання і швидкість ультразвуку у монокристалах LiF. *О.М.Петченко, Г.О.Петченко, С.М.Бойко, А.С.Литвиненко.*

Імпульсним методом на частоті 7,5 МГц досліджено взаємозалежність дислокаційного поглинання і швидкості ультразвуку у монокристалах LiF із залишковою деформацією ε = 0,65 % за T = 300 К в інтервалі доз опромінення 0–1057 рентген. Встановлено, що за умов опромінення кристалів рентгенівськими променями істотно збільшується концентрація радіаційних центрів, що закріплюють легкорухливі дислокації. У результаті обмеження числа вказаних дислокацій згасання ультразвуку стрімко зменшується, а швидкість його поширення у зразку збільшується.

1. Introduction

As we can find out from the reviews [1, 2], quite a lot of experimental work has been devoted to the study of the radiation exposure effect on crystal properties. From the previous studies follows that an increase of radiation damages' number in alkali halide crystals stimulates the occurrence of optical, acoustic and other effects caused by the interaction between these point defects and mobile dislocations. Taking into account that the beginning of the settling process on various pinning points' dislocations (including centers of radiation origin) recording is reliable, then the use of acoustic methods is the most effective [1]. With their help, it is possible to quite reliably record the slightest changes in the average effective length of a dislocation segment L, oscillating in the field of an ultrasonic wave, which, by virtue of law $\alpha \sim L^4$ [1, 3], very significantly affect the measured ultrasound attenuation α in the sample.

From the review [1], one can learn that in early researches, when studying radiation effects, low-frequency methods of amplitude-dependent internal friction were used most often.

These methods are very sensitive to the appearance of radiation defects in the crystal, but their application presents certain difficulties. When a low-frequency ultrasonic wave of large amplitude is transmitted through the sample under study, deformation in it can occur directly by ultrasound. As a result, not only the value of L can change, but also the dislocation density Λ, which in experiments on the effects of low radiation usually remains unchanged.

Moreover, with such amplitudes of ultrasonic waves, small portions of radiation point defects cannot have a noticeable pinning effect on a moving dislocation. Large doses' irradiation of crystals (more than 10^3 R) is used (to obviously display its stopping action) in low-frequency experiments. This exposure, according to [4], can lead to a change in the mechanical characteristics of crystals, as well as stimulate the emerging of optical absorption bands [5–7].

In this connection, the need arises to use such experimental methods that would be able to detect the occurrence of radiation defects at the initial stages of crystals' irradiation. Such methods include the amplitude-independent, pulse-type echo method of the high frequency range [8–43], which is notable for its high information content and high sensitivity to the impact on the dislocation of weak stoppers.

The works [8–43] convincingly demonstrate the high sensitivity of the method [1] to subtle structural changes in the crystal. It should be noted, however, that the mentioned works realize the capabilities of the model [3] only partially. They have a common direction which means that they study the phonon-dislocation interaction and the interaction of mobile dislocations in a crystal with Mott and Friedel-type stoppers [3] on the basis of the frequency spectra processing of the dislocation decrement of ultrasonic attenuation $\Delta_d(f)$ in the dislocation's resonance region and post-resonance one.

As the authors of the string model [3] pointed out, this kind of experiment is very informative and gives us the most correct results regarding crystals' under study dynamic and structural characteristics' research, since the conditions of the experiment most fully meet the conditions and limitations of the model [3]. However, the authors of [3] also proposed a slightly different algorithm for finding the above-mentioned crystal characteristics, which is based on the results of studying the speed and ultrasound absorption in the pre-resonance region of the $\Delta_d(f)$ curves.

There are very few examples of its practical implementation [1, 3, 8]. The fact is that for processing the experimental data according to the indicated algorithm, it is necessary to have data on the resonance region and the descending branches of the $\Delta_d(f)$ curves, which are very difficult to obtain due to the absence of standard experimental equipment for such tasks. The notorious difficulty also limited the appearance of new works on the dynamic and structural parameters' research in a crystal from experiments on the ultrasonic velocity dispersion [31]. The possibility to process obtained experimental data is also offered by the model [3].

The above reasoning shows that in the presence of the works' array [29–43], where the structural changes in crystals were studied in three ways — by the resonance region, by the descending branch of the $\Delta_d(f)$ curves, and by using dispersion experiments. Of particular interest is the practical implementation of the model [3] for the region of low (pre-resonant) frequencies. The presence of results in this area would allow us to generalize and talk about a complete check of all the possibilities that the model [3] opens up for experimenters.

This consideration's become the starting point for this work.

2. Experimental

In this work, we studied the effect of small X-ray radiation doses on the dislocation absorption and ultrasound velocity in LiF single crystals with a residual deformation of 0,65 % at a frequency of 7.5 MHz at a temperature of 300 K. The specimens with a purity of 10^{-4} wt.%, with the crystallographic orientation <100>, and $17\times17\times27$ mm^3 in dimension we used carrying out the experiments. According to the technology described in works [29–43], the specimens to study, after their cutoff, were finely polished to achieve the working sur-

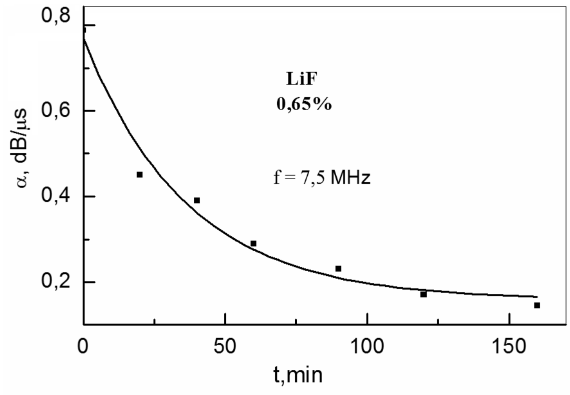

Fig. 1. Dependence of the dislocation's ultrasound attenuation on the irradiation time for deformed (0.65 %) LiF crystals at T = 300 K.

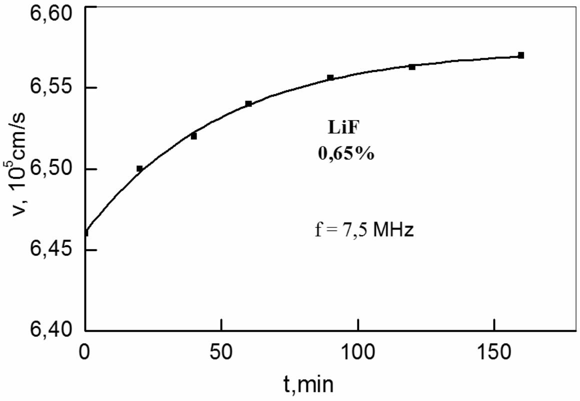

Fig. 2. The dependence of the elastic waves' velocity on the irradiation time for LiF crystals with a residual deformation of 0.65 % at T = 300 K.

faces' non-parallelism of approximately ±1 μm/cm, which was controlled by means of an IKV optimeter. The surface non-parallelism in the system "piezoquartz-sticker-specimen" could also be estimated independently when imposing the exponential reference signal on a series of reflected pulses observed on the oscilloscope screen in the course of crystal sounding. The ultrasound attenuation was measured by applying a calibrated exponent to the echo signals, and the pulse interference method and the selective method were used to measure the velocity on the equipment described in [29–43]. To remove the internal stresses that could emerge owing to a mechanical treatment of the specimens, the latter were annealed in a muffle furnace MP-2UM for ~ 12 h at a temperature of about $0.8T_{mp}$ (T_{mp} — melting point) and, then, slowly cooled down to room temperature. For highly mobile dislocations to be introduced into the crystal, the latter was preliminarily deformed to achieve the residual strain ε = 0.65 %. The specimens were deformed by squeezing them on an Instron tensile machine at a rate of about 10^{-5} s^{-1}. In this regime of deformation [29–43], no slip bands arise, and the etch pits regularly cover the crystal surface, which enables the dislocation density Λ to be accurately determined with the use of the software Photoshop. The technology for isolating the dislocation component from the total absorption was the same as in [29–43]. The standard installation of URS-55 was used to X-ray irradiation the studied LiF single crystals. The X-ray irradiation dose power at the location of the studied crystal, according to our estimates, was ~ 0.11 R/s. To avoid the appearance of irregularities in the sample caused by the action of internal mechanical stresses [2], each of the 4 side faces parallel to the long axis of the crystal was irradiated alternately for 20 min, which corresponded to a total dose of 1057 R.

3. Results and discussion

There were studied the dislocation's attenuation α dependence on the time of X-ray irradiation for LiF samples with residual deformation ε = 0.65 %, and Fig. 1 shows the obtained results. It can be seen that already after the first 20 min of the irradiation the ultrasound attenuation in the sample declined by 0.34 db/μs, and during the remaining measurement time its value decreased by another 0.3 db/μs.

Fig. 2 presents the experimental data obtained on the same sample, when measuring the velocity of ultrasound, and turned out to be quite expected. It is evident from the above-mentioned figure that with an increase in the dose of irradiation, the effect of ultrasonic wave propagation velocity growth in the crystal is manifested to a greater extent.

It should be noted that a similar effect of changes in acoustic characteristics was previously observed in study [8] when NaCl crystals were irradiated. According to the author of [8], this is due to a decrease in the effective length of the dislocation loop L, due to its fixing with radiation defects, the number of which is added during the irradiation time to some dislocation loop with the initial length L_0. In accordance with the work [3], after irradiation for a time t, the length of the dislocation segment is equal to:

$$L = \frac{L_0}{1 + c(t)}, \quad (1)$$

where $c(t)$ is the concentration of point defects. According to the theory [3], for the low-frequency region related to the ascending branch of a damped dislocation resonance, the expression for the attenuation coefficient of ultrasound has the form:

$$\alpha = K_1\omega^2\Lambda L^4, \quad K_1 = 8.68 \cdot 10^{-6}\left(\frac{4Gb^2}{\pi^4 C}\right)\left(\frac{B}{\pi^2 C}\right), \quad (2)$$

where ω is the cyclic frequency, G is the shear modulus of the acting slip system, b is the value of the Burgers vector, C is the effective tension of the bent dislocation, B is the damping constant.

Using the relation (1), we can obtain an expression for attenuation in this form:

$$\alpha = \alpha_0\left[\frac{1}{1 + c(t)}\right]^4, \quad (3)$$

where $\alpha_0 = K_1\omega^2\Lambda L_0^4$ — initial attenuation before irradiation.

Based on equation (3), the concentration dependence $c(t)$ on the irradiation time t takes the form:

$$c(t) = \left(\frac{\alpha}{\alpha_0}\right)^{-\frac{1}{4}} - 1. \quad (4)$$

It can be seen that the function $c(t)$ is expressed through the initial attenuation at $t = 0$ and the attenuation at an arbitrary time t.

Similarly, the dependence $c(t)$ can be calculated from measurements of the velocity of ultrasound. From the theory [3] it follows that:

$$\frac{\Delta\nu}{\nu_0} = K_2\Lambda L^2, \quad K_2 = \frac{4Gb^2}{\pi^4 C}. \quad (5)$$

Relation (5), taking into account (1), allows us to obtain an expression for concentration in the form:

$$c(t) = \left[\frac{\Delta\nu/\nu_0}{(\Delta\nu/\nu_0)_0}\right]^{-\frac{1}{2}} - 1, \quad \left(\frac{\Delta\nu}{\nu_0}\right) = K_2\Lambda L_0^2, \quad (6)$$

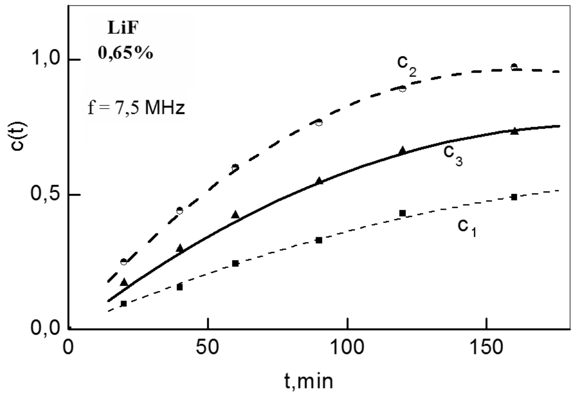

Fig. 3. The dependencei of point defects' concentration on the time of irradiation in LiF crystals with a residual deformation of 0.65 % at T = 300 K.

where $\left(\frac{\Delta\nu}{\nu_0}\right)$ and $\left(\frac{\Delta\nu}{\nu_0}\right)_0$ — relative changes in the speed of sound at irradiation time t and t = 0, respectively.

Fig. 3 shows the course of curves $c_1(t)$ and $c_2(t)$ calculated by formulas (4) and (6), respectively, as well as the average value of $c_3(t)$, (where $c_3 = 0.5(c_1 + c_2)$.

It can be seen that the dependence of concentration on irradiation time is not linear. However, according to the authors of [1, 3], it can still be described in the first approximation by the equation:

$$c(t) = \frac{nrL_0}{\Lambda}t, \quad (7)$$

where n is the number of fixing centers in 1 cm^3 generated by radiation with a dose rate of 1 R per unit of time, r is the irradiation dose power, Λ/L_0 the number of dislocation loops in 1 cm^3, and (nrL_0/Λ) the number of additional pinning centers added on average to each dislocation loop L_0 per time unit.

If, as a rough approximation, the curve $c_3(t)$ shown in Fig. 3 is approximated by a linear dependence $c(t) = \beta t$, then for the initial part of this curve the coefficient is 0,05. Using [39] the values for dislocation density $\Lambda = 1.74{\cdot}10^6$ cm^{-2}, determined by the etching points method, and the length of the dislocation segment $L = 8.35{\cdot}10^{-5}$ cm, calculated from the parameters of the dislocation resonance, as well as the value of $r = 6.6$ R/min $n = 1.57{\cdot}10^8$ cm^{-3} was found.

Analyzing the results of this work, we found that the concentration of radiation defects of fixing dislocations in LiF and a similar value $n = 3{\cdot}10^8$ cm^{-3}, obtained by irradiating NaCl crystals [8], the dose rate of which was 58 R/min, were almost the same.

Using the value $n = 1.57{\cdot}10^8$ cm^{-3} found in this work, we were able to calculate the concentration of the pinning centers created in the crystal during its irradiation for 160 min, which turned out to be $2.5{\cdot}10^{10}$ cm^{-3}. During this time, as shown by the course of the experimental curves (Figs. 1 and 2), highly mobile dislocations are effectively fixed by stoppers, which are the pinning centers of X-ray nature. Due to X-ray processing, the crystal under investigation restores its original acoustic performance, which it had immediately after preparation and annealing.

Comparing our concentration results with the data of [6,7], obtained on the same crystal by an optical absorption method using Smakula's theory [44], it was found that this method yields a value for the *F*-center concentration of $10.85{\cdot}10^{15}$ cm^{-3}, which differs markedly from the value of n, found by the acoustic method [3]. This difference may be due to the fact that in alkali halide crystals, pinning centers arise directly on dislocations [11, 29–43]. This means that the concentration of point defects, which ensures the complete fixation of mobile dislocations, is noticeably less than the volume concentration of color centers, which can be detected by the optical absorption method [5–7, 44–50].

4. Conclusions

The effect of X-ray irradiation in the dose range 0-1057 R on dislocation absorption α and the ultrasonic velocity ν in LiF single crystals with a residual deformation of 0.65 % measured at 7.5 MHz at 300 K is studied.

The experiments gave us the opportunity to reveal that already after the first t = 20 min of irradiation, the ultrasound attenuation in the sample decreases sharply, and the speed of ultrasonic wave propagation increases significantly. The analysis showed that these changes in acoustic characteristics are associated with a decrease in the effective length of the dislocation segment L due to its pinning by radiation defects.

Based on the processing of the experimental data obtained in the framework of the Granato-Lucke theory, the concentration of centers, pinning the dislocations that appeared in the crystal upon irradiation was calculated. During the time of radiation exposure, as shown by the course of the curves $\alpha(t)$ and $\nu(t)$, easily mobile dislocations are effectively fixed by radiation stoppers, as a result of which the crystal under study almost completely restores its original acoustic characteristics.

The obtained concentration results were compared with the data received on the same crystals by means of the optical absorption method using Smakula's theory. The optical method was found to give a value for the concentration of *F*-centers, which differs markedly from the value n found by the acoustic method. Such a difference is possible due to the fact that in ionic crystals the pinning centers arise directly on dislocations. This means that the number of defects ensuring the complete fixation of mobile dislocations should be significantly less than the volume concentration of color centers, which can be determined by the method of optical absorption.

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